

Amendments to the Specification:

On page 1, after the title, insert the following new paragraph:

CROSS-REFERENCE TO RELATED APPLICATION

This application claims priority to PCT Appln. No. PCT/EP2004/004564 filed April 29, 2004, and to German application 103 21 942.0 filed May 15, 2003.

At page 1, line 3, please add the following heading and subheading as shown below:

BACKGROUND OF THE INVENTION

1. Field of the Invention

At page 1, line 12, please add the following subheading as shown below:

2. Description of the Related Art

At page 2, line 25, please add the following heading as shown below:

SUMMARY OF THE INVENTION

At page 2, line 26, please amend the following paragraph as shown below:

It is an object of the invention to provide a process by which ethylene-vinyl alcohol copolymers are obtainable in powder form in a simple manner and with high purity. These and other objects are achieved by precipitation of the copolymer from its alcoholic hydrolysis medium by cooling with a temperature gradient, optionally with the addition of water.

At page 2, line 31, please add the following heading as shown below:

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

At page 2, line 31, please amend the paragraph as shown below:

The invention provides a process for preparing pulverulent ethylene-vinyl alcohol copolymers by free-radical polymerization of ethylene and one or more vinyl esters, and optionally further monomers copolymerizable therewith, and subsequent hydrolysis of the thus obtained ethylene-vinyl ester copolymers to give ethylene-vinyl alcohol copolymers, characterized in that the ethylene-vinyl alcohol copolymer, after the hydrolysis, is precipitated from alcoholic solution by means of cooling with a temperature gradient, and optional addition of water, the temperature gradient in the case of ethylene-vinyl alcohol copolymers ~~which derive~~ derived from low molecular weight ethylene-vinyl ester copolymers having a weight-average molecular weight Mw of from 2000 to ~~100,000~~ 100,000 g/mol being from -0.1°C/min to -10°C/min, and the temperature gradient in the case of ethylene-vinyl alcohol copolymers which derive from high molecular weight ethylene-vinyl ester copolymers having a weight-average molecular weight Mw of > 100 000 g/mol being from -0.1°C/min to -1°C/min.

At page 3, line 36, please amend the paragraph as shown below:

To obtain relatively low molecular weight products, i.e. having a weight-average molecular weight Mw of from 2000 to ~~100,000~~ 100,000 g/mol, preferably from 5000 to ~~60,000~~ 60,000 g/mol, preference is given to using relatively high amounts of initiator, ~~of~~ preferably from 0.2 to 1.0% by weight, more preferably from 0.4 to 0.8% by weight, based in each case on the total weight of the comonomers without the ethylene fraction. In order to obtain relatively high molecular weight products, i.e. products having a weight-average molecular weight Mw > ~~100,000~~ 100,000 g/mol, preference is given to polymerizing with an amount of initiator of from 0.01 to 0.1% by weight, based on the total weight of the comonomers without the ethylene fraction.

At page 8, line 25, please amend the paragraph as shown below:

In the case of ethylene-vinyl alcohol copolymers which ~~are derive~~ derived from low molecular weight ethylene-vinyl ester copolymers having a weight-average molecular weight Mw of from 2000 to ~~100-000~~ 100,000 g/mol, preferably from 5000 to ~~60-000~~ 60,000 g/mol, the temperature gradient is from -0.1°C/min to -10°C/min, preferably from -1°C/min to -10°C/min.

At page 8, line 33, please amend the paragraph as shown below:

In the case of ethylene-vinyl alcohol copolymers which ~~are derive~~ derived from high molecular weight ethylene-vinyl ester copolymers having a weight-average molecular weight Mw of > ~~100-000~~ 100,000 g/mol, the temperature gradient is from -0.1°C/min to -1°C/min, preferably from -0.1°C/min to -0.5°C/min.

At page 9, line 31, please amend the paragraph as shown below:

The precipitated ethylene-vinyl alcohol copolymer can be removed by means of filtration and is generally present with a residual moisture content of $\leq 50\%$. The product is typically dried, for example with hot air, and is present in powder form after the drying. The particle size, determined as the mean volume diameter D_v , is from 20 to 2000 μm , preferably from 100 to 1000 μm . The desired particle size can be controlled by the process, i.e. via the temperature gradient in the cooling and precipitation, via the addition of water and its amount, and via the stirrer speed in the crystallization. The complex melt viscosity of the products is preferably from 0.5 to ~~100-000~~ 100,000 Pas, in particular from 1 to 1000 Pas (at 180°C; oscillating measurement at 1 Hz with plate/plate test system, instrument: Bohlin CVO 120 HR).

At page 11, line 14, please amend the paragraph as shown below:

It is possible to obtain low molecular weight products for which the range of the molecular weight can be very large. The low molecular weight ethylene-vinyl ester copolymers ($M_w \leq \text{100-000 } \underline{100,000} \text{ g/mol}$), particularly the very low molecular weight ethylene-vinyl ester copolymers having $M_w \leq \text{50-000 } \underline{50,000} \text{ g/mol}$, can be converted by the process to readily processible, pulverulent ethylene-vinyl alcohol copolymers. The resulting products have a homogeneous structure. The low solution viscosity leads to advantages in the polymerization: it is still possible to stir solutions having a solids content of up to 95%.

At page 12, line 35, please amend the paragraph as shown below:

Analyses:

Solids content SC: 30.55% (in methanol);

Viscosity (Höpler – 10% in ethyl acetate): 14.34 mPas;

Acid number AN (methanol): 5.61 mg KOH/g;

Residual vinyl acetate: 55 ppm; hydrolysis number: 519.05 mg KOH/g;

Ethylene content from ^1H NMR: 20.69% by weight (44.54 mol%);

SEC $M_w = \text{114632 } \underline{114,632}$, $M_n = \text{35223 } \underline{35,223}$, polydispersity = 3.52 (NB: eluent is always THF; the data are relative based on polystyrene standards);

Tg of the dried resin: 0.9°C. N.B.: the Tg of ethylene-containing solid resins can be estimated very simply by the following formula (Fox equation): $1/T_g = \% \text{ by wt. (Et)}/180K + \% \text{ by wt. (VAc)}/316K$. A theoretical value of 0.13°C is calculated here.

At page 13, line 36, please amend the paragraph as shown below:

Analyses:

SC: 45.55% (in methanol); viscosity (Höpler – 10% in ethyl acetate): 2.23 mPas; AN (methanol): 5.05 mg KOH/g;

Residual vinyl acetate: 16 ppm; hydrolysis number: 425.37 mg KOH/g;

Ethylene content from ^1H NMR: 30.48% by weight (57.44 mol%);

SEC $M_w = \text{23666 } \underline{23,666}$, $M_n = 8900$, polydispersity = 2.66;

Tg of the dried resin: -17.0°C.

N.B.: a theoretical value of -16.3°C is calculated here.

At page 21, line 29, please amend the paragraph as shown below:

Example 7 (very low molecular weight EVOH):

In a 3-liter jacketed, heatable glass reaction vessel (with distillation head and condenser, and dropping funnel), 653.6 g of a 76.5% methanolic solid resin solution of an ethylene-vinyl acetate copolymer which has been obtained analogously to example B, contains 70.9 mol% of ethylene and 29.1 mol% of vinyl acetate, and has an average molecular weight Mw of 6872 g/mol were weighed in. Subsequently, the solution was heated with stirring (200 rpm, blade stirrer) to a jacket temperature of 60°C. Once the temperature had been attained, the hydrolysis process was started by dropwise addition of 820 g of a 6.1% by weight methanolic sodium hydroxide solution, corresponding to 50 g of solid NaOH = 10.0% of the amount of solid resin. The metered addition was effected by twice adding alkali for 15 minutes each at an interval of 30 minutes.

At page 23, line 28, please amend the paragraph as shown below:

Example 9 (very low molecular weight EVOH):

In a 3-liter jacketed, heatable glass reaction vessel (with distillation head and condenser, and dropping funnel), 1091.4 g of a 73.3% methanolic solid resin solution of an ethylene-vinyl acetate copolymer which has been obtained analogously to example B, contains 67.6 mol% of ethylene and ~~32.5~~ 32.4 mol% of vinyl acetate, and has an average molecular weight Mw of 5520 g/mol were weighed in. Subsequently, the solution was heated with stirring (200 rpm, blade stirrer) to a jacket temperature of 80°C. Once the temperature had been attained, the hydrolysis process was started by adding dropwise 920 g of an 8.7% by weight methanolic sodium hydroxide solution, corresponding to 80 g of solid NaOH = 10.0% of the amount of solid resin. The alkali was metered in within one hour. During the hydrolysis time of 2.5 hours, 821 g of methyl acetate/methanol solvent mixture were removed under reduced pressure

and 342 g of methanol were added. After this time, the hydrolysis was terminated by addition of 25 ml of glacial acetic acid, i.e. neutralization was effected to $\text{pH} = 7.2$.